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ASSESSMENT OF PRODUCTION VARIABLES ON THE PERFORMANCE OF CERAMIC MEMBRANES

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ABSTRACT

Water crisis is one of the most important topics nowadays. The water crisis is approaching due to several factors, which is why finding alternative sources of water is important. Water treatment has gained its importance as it decreases the amount of waste water in addition to producing water that can be used in different applications based on its characteristics. Several water treatment techniques are present nowadays; however, research is needed to lower the cost of such techniques in addition to increasing productivity and maximize their range of applicability. Several water treatment techniques are capable of producing fresh water from different types of feed water (saline water, waste water, etc.), these techniques include membrane technologies and thermal technologies. Due to high energy consumption, and lower productivity in thermal techniques membrane techniques are mostly used nowadays in the applications of water treatment. Ceramic membranes have attracted researchers due to their characteristics and wide range of applicability. In this paper, different ceramic membrane types are illustrated; production techniques and the effect of each production variable on the membrane characteristics are shown, in addition to applying a comparison between different additives. Finally, a comparison of performance is carried out.

Keywords: Ceramic membranes, Coating methods, Fabrication techniques, Thermal treatment, Water treatment.

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1. INTRODUCTION

Finding alternative sources of water is crucial since the current water crisis is one of the most essential concerns. Water treatment has gained its importance as it reduces the quantity of wastewater, besides providing water that may be utilized in a variety of applications. There are many water treatment procedures available today, but more research is needed to reduce the cost of such techniques to increase productivity and minimizing their negative impact.

2. WATER CRISIS

Water Crisis is considered one of the most important topics nowadays, the world health organization stated that by 2025 half of the world's population will be living in a water stressed areas [1], there are several factors affecting the water available per capita as shown below:

2.1 Population Explosion

As long as the population in a certain area increases, the water available per capita decreases, as a result finding alternative resources of fresh water is needed so as not to reach the water deficit. The current population growth rate is 1.05% per year, the current world population is around 8 billion and it is anticipated that the world population will reach 10 billion in 2057. Egypt is ranked 14 in the top 20 countries in the world by population, the population exceeded 100 million in 2020 with a growth rate of about 2% and a population density of 103 people per km^2 which is considered a high population density as shown in Figure-1 [2].

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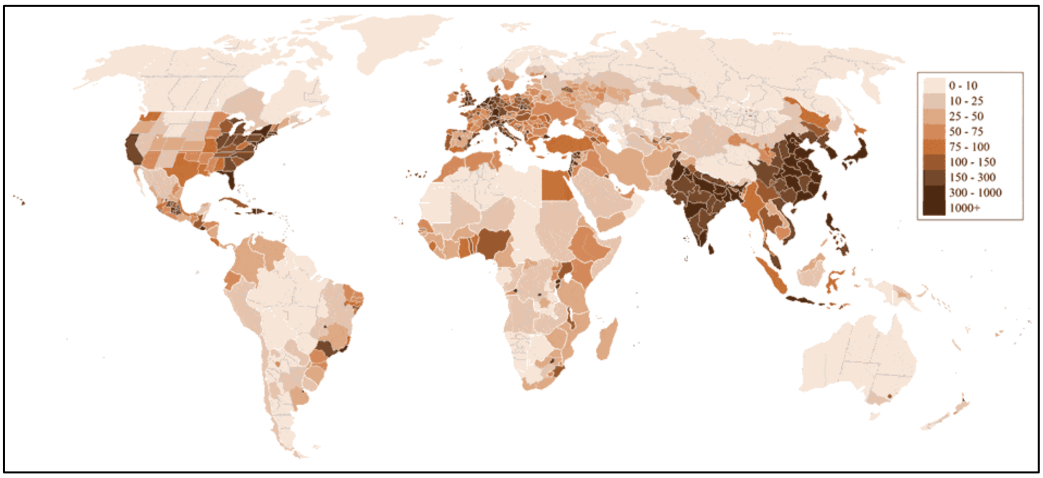

Figure-1. World population density (people/km^2) [2].

2.2 Water Pollution

Water pollution is one of the major factors that affect the water availability in a region, sources of fresh water can be contaminated by different types of waste including industrial waste, agricultural waste, etc., and due to contamination, water will not be eligible for direct use and further treatment will be needed. In Egypt, the Nile is considered the main source of fresh water, there is a total population exceeding 20 million living by the Nile who are dumping their untreated sewage in it, which affects the quality of the water badly [3]. On the other hand, the industrial and agricultural pollution to the Nile, it was reported by the Egyptian Environmental Affairs Agency that around 19 billion m^3 of wastewater is dumped in the Nile per year, 71% of which is agricultural waste mixed with sewage [4].

2.3 Inefficient Irrigation

Inefficient irrigation consumes a huge amount of water, thus affecting the fresh water available for human use, new irrigation techniques can reduce the amount of water used in different agricultural sectors. In Egypt, only 6% of the area is considered arable and the rest is desert, using outdated techniques for irrigation is considered one of the most water-wasting categories such as flood irrigation. Nearly 3 billion cubic meters of water are lost per year due to evaporation as most of the adjacent farms to the Nile use canals and sub canals to deliver water from the Nile to the farms [5].

2.4 Regional Upheavals

Regional upheavals affect the amount of water available in a country, such as building dams, and as a result, the amount of water available in a country is affected badly. In Egypt, one of the biggest challenges nowadays is encountering a shortage in dry months due to the dam which is built in Ethiopia, as this dam would affect the water available in Egypt, especially during the filling time [5].

As a result of these factors, water treatment is needed to be able to increase the amount of fresh water available for human use.

3. WATER TREATMENT

There are several water treatment techniques that are capable of producing fresh water from several types of feed water (saline water, waste water, etc.), these methods include membrane technologies and thermal technologies [6].

Thermal technologies include multi-stage flash distillation, multi-effect distillation, and vapor compression. On the other hand, membrane technologies include nano filtration and reverse osmosis. However, due to high energy consumption, and lower productivity in thermal techniques the membrane techniques are mostly used nowadays in the applications of water treatment [7].

3.1 Membrane Water Treatment Techniques

A membrane is considered a barrier that separates components by restricting the flow of components selectively. The membranes can be classified according to deriving force as shown in the following table [8].

Table-1. Membrane technology classification according to the force applied [8].

Driving Force	Examples
Pressure driven	• Microfiltration • Ultrafiltration • Nano-filtration • Reverse osmosis.
Concentration gradient	• Dialysis
Temperature driven	• Membrane distillation
Electrical potential	• Electrodialysis

Pressure driven techniques are the most widely used in water treatment due to their low operating cost compared to other techniques in addition to high flow rates of product. Each membrane has its range of applications as illustrated below [9,10].

a) Microfiltration (0.1-5 µm), can be used to separate suspended solids and bacteria.

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b) Ultrafiltration (1-100 nm), can be used to separate suspended solids, bacteria, and macromolecules.
c) Nanofiltration (0.5 -10 nm), can be used to separate suspended solids, bacteria, macromolecules, and multivalent ions.
d) Reverse osmosis (less than 0.5 nm), can be used to separate suspended solids, bacteria, macromolecules, and multivalent and monovalent ions.

3.2 Membrane Technology

A membrane is a layer that allows elements to pass selectively, membrane technology is used in many sectors including pharmaceutical, food, waste water treatment, etc., and membranes can be classified into ceramic and polymeric membranes. Polymeric membranes have the advantage of low production cost and small pore size distribution; however, they cannot be used in high temperature applications, besides not being suitable to be used in acidic environments. On the other hand, ceramic membranes are stable in high temperatures in addition to being suitable to be used in different chemical conditions and have a longer lifetime [11]. The advantages and disadvantages of each membrane technology been illustrated in the following table.

Table-2. Advantages and disadvantages of polymeric membranes versus ceramic membranes [12,13].

	Polymeric membranes	Ceramic membranes
Advantages	Low cost	Low fouling High temperature stability Resistance to chlorine
Disadvantages	High fouling Low temperature stability Lower lifetime compared to ceramic membranes	High cost Needs careful handling

Ceramic membranes are considered one of the promising topics for research and development, finding alternative raw materials and manufacturing conditions are important to lower their cost of production thus giving them a huge value versus the polymeric membranes.

3.3 Ceramic Membranes

Ceramic membranes consist of three main layers, the support layer, intermediate layer, and selective layer as shown in Figure-2, the support provides the needed strength for the membrane, the intermediate layer provides the selectivity needed by the membrane and the selective layer provides the separation objective [12].

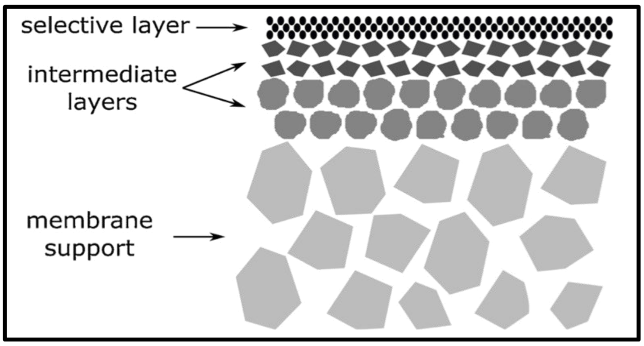

Figure-2. Different layers in ceramic membranes [12].

3.3.1 Types of ceramic membranes

Ceramic membranes can be classified according to structure or geometrical configuration, their structure could be dense or porous, and their geometry could be plate and frame, tubular, capillary, or hollow fiber [14].

A. According to Structure

- Dense membranes are used in gas separation and their separation technique is that selected molecules can dissolve in the membrane then diffuse and desorbs from it.
- Porous membranes are used for solid liquid and solid gas separation according to their pore size, the structure could be symmetric if the pores are equally sized and asymmetric if it decreases from the bottom to the surface.

B. According to Geometry

- Plate and frame membrane has a pillow shape that two membranes form when they are inside a membrane unit, multiple pillows are placed in the module with a spacing that depends on the dissolved solids present in the water, and the flow of water is inside to out, so the permeate is collected from the spaces.
- Tubular membranes are membranes that have a straw shape that can be used with water without pre-treatment. This kind of membrane is used inside a tube housing, and the water is pumped into the core so the permeate can pass inside to out and can be collected from the tube.
- Capillary membranes are equivalent to tubular however they have lower diameters and the flow can be from inside to out and vice versa. The advantage of this type of membrane is that they have a high packing density.
- Hollow fiber membranes are membranes that can be used with water that has low suspended solids content, they have high packing density and are commonly used in reverse osmosis and nano filters, water passes in the core and the permeate is collected from the surrounding of the membrane.

3.3.2 Fabrication of ceramic membranes

To prepare low-cost ceramic membranes, each step in the manufacturing of the membrane must be altered to reduce its cost. Manufacturing of ceramic membranes has three main stages, selection and preparation of raw material, preparation method (shaping), and thermal treatment.

A. Raw Materials and Additives

First of all, low-cost raw materials must be selected such as kaolin clay in addition to the usage of industrial wastes such as fly ash and rice husk [15][16], in this way the raw material cost will be reduced thus reducing the final production cost of the membrane. The preparation of the raw material includes grinding to achieve the particle size that can be mixed to result in a membrane with the needed specifications.

Commonly commercialized membranes use alumina as it is stable thermally and chemically, in addition to using kaolin as it gives the membrane several benefits such as high refractory properties, and hydrophilic behavior [13]. The following figure shows the most used raw material to produce low-cost ceramic membranes.

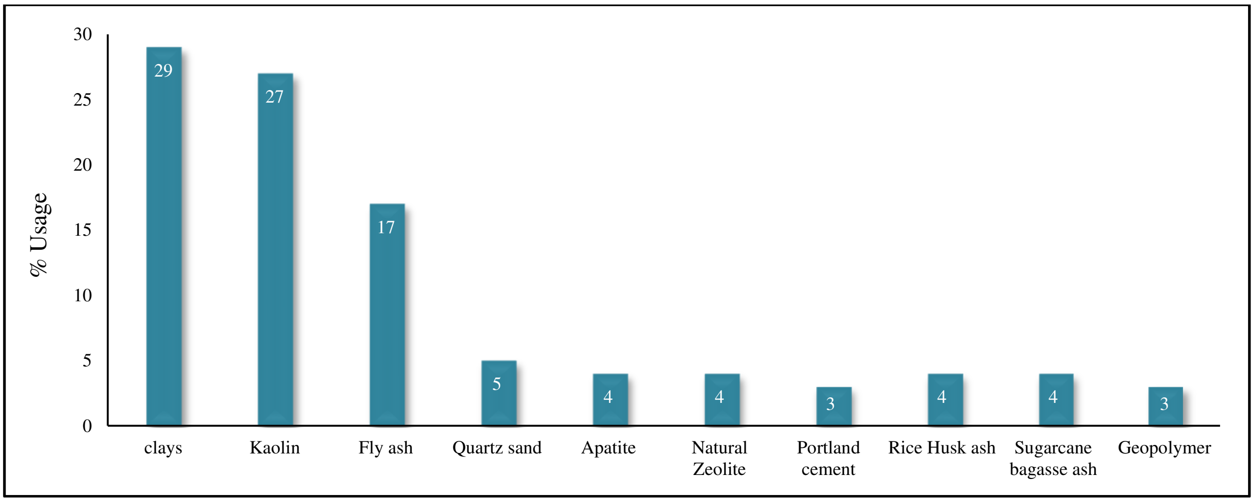

Figure-3. Usage percentage of different raw materials in ceramic membranes [13].

Additives can be added to the membrane to enhance its properties, such as zeolites, which add adsorption abilities to the membrane. For example:

- Zeolites are known for their superior ammonia ion absorptivity, which is advantageous for applications in the treatment of fertilizer-contaminated water.
- Apatite is not only able to efficiently adsorb metal contaminants but is also effective in the removal of anionic and cationic dyes by adsorption.

It was found that the cost of ceramic membranes could range from 2$ up to 130$ per m^2 based on the material used, the following table shows different raw materials and the cost of membranes produced [17].

Table-3. The influence of raw material on the cost of membranes [17].

The Material Used for the Preparation of Membrane	Cost of Raw Material (USD) /m^2
Fly ash and titania	2
Fly ash quartz and calcium carbonate	5
Kaolin, ball clay, feldspar, calcium carbonate, and pyrophyllite	10
Fly ash, calcium carbonate, sodium carbonate, and boric acid	17
Clay, sodium metasilicate, sodium carbonate, and boric acid	19
Fly ash, quartz, calcium carbonate, and titania	25
Kaolin, quartz, calcium carbonate, sodium carbonate, boric acid, sodium metasilicate, and polyvinyl alcohol	78
kaolin, quartz, calcium carbonate, sodium carbonate, boric acid, and sodium metasilicate	130

Pore forming Agents are used to increase the porosity and permeability of the membranes [18]; different materials were used such as different types of starch [19], saw dust [20], and so on. Porosity is the percentage of the volume of void spaces in the membrane, porosity increases with increasing the quantity of pore forming agents, on the other hand, increasing the quantity of pore forming agents excessively would result in several drawbacks such as a decrease in the strength of membrane in addition to increasing the shrinkage. The decrease in the membrane strength is due to the elimination of pore forming agent in addition to the presence of void spaces in the membrane, on the other side, a zero-pore forming membrane size is reduced due to the elimination of water only, but after using pore forming agents not only the water is eliminated but also the pore forming agents which increase the shrinkage [21].

B. Preparation Methods

Secondly the preparation method, there are two common ways to produce ceramic membranes, flat shape circular disk membranes, and tubular membranes, each method will affect the geometry, microstructure, and specifications of the membrane.

a. Powder pressing

The raw materials are mixed with additives such as binders, pore forming, etc., then it is pressed to get the desired shape. Finally, drying and sintering take place to produce a membrane with suitable properties. This method is cost effective and preferred for high volume production [22].

b. Extrusion

In this method raw materials and additives are mixed to get a viscous paste, then an extruder is used to get the membrane with the desired shape. Finally, drying and sintering take place to produce a membrane with suitable properties. This method has the advantage of producing membranes with high mechanical strength and high surface to volume ratio [11].

c. Slip casting

Slip casting is used as it has a lower cost than other techniques besides being simple, in which the slurry is poured into a microporous mold which draws out the fluids and keeps a layer of membrane. The drawbacks of this technique are that the control of the thickness of the membrane is hard, and it requires a long time [13].

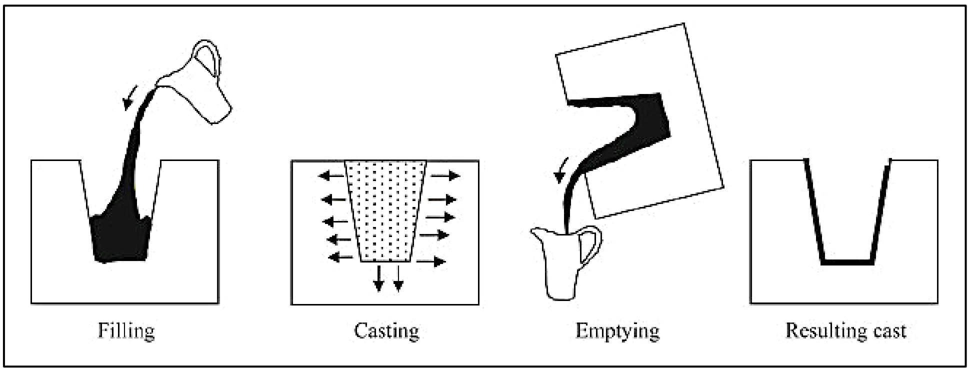

Figure-4. Slip casting fabrication method [23].

d. Thermal treatment

The third step is thermal treatment, it is one of the most important steps in the manufacturing of the membrane; as in this step the membrane gets its strength and structure. The rate of both heating and cooling is kept below 5 C/min to lower thermal stresses, first of all, the membrane is dried to remove the water content, and then the temperature of the sintering process is chosen below the melting point of the raw material. The sintering is performed in cycles to perform a selective elimination of additives. For example, at when the membrane is kept for some time at 100-110°C, the elimination of water takes place, at 250°C elimination of organic additives takes place, at 500°C, the elimination of saw dust takes place [24], and so on. Figure-5 illustrates the thermal treatment cycle.

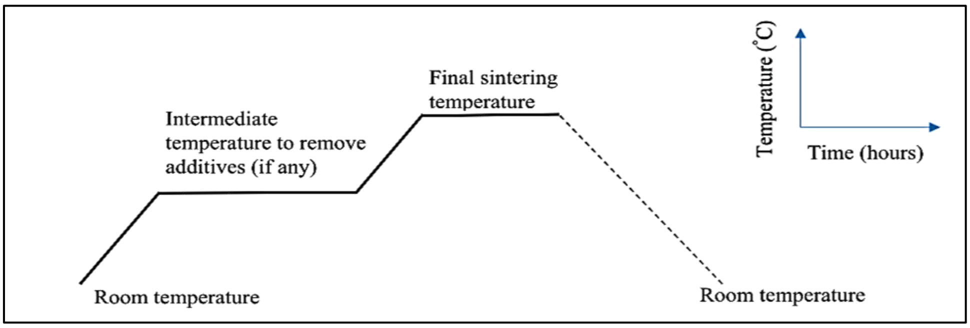

Figure-5. Thermal treatment cycle [24].

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Choosing the sintering temperature is crucial as it affects the pore size, permeability, crystal size, mechanical strength, and chemical resistance. The effect of increasing the sintering temperature is tabulated below:

Table-4. Effect of increasing sintering temperature on different properties [25].

Properties	Effect of increasing sintering temperature	Justification
Pore size	increases	Due to forming large pores and elimination of small pores [26–28]
permeability	increases	Due to the formation of larger pore sizes [29]
Mechanical strength	Increases	Due to the formation of low crystal size which leads to densified membranes [30,31]
Chemical resistance	Increases	Based on laboratory studies. [32]

a) Coating

Applying a coating layer controls the pore size of the membrane; as a result, it improves the selectivity. Coating can be applied using different and choosing the most suitable method is crucial as it affects the performance of the membrane [25], different methods as illustrated below:

- **Spray method**

In this method the coating is sprayed on the support, this method can reduce particle penetration and it has several factors such as the pressure, distance, and dry time between cycles [33], which can result in 60 to 200 microns [25].

- **Dip coating method**

In this method the membrane is dipped in the coating suspension, the viscosity of the suspension and the time of coating are considered crucial factors in this method, after which it is dried and calcinated. This method can be used to perform multiple coatings, in which after calcination of the first layer, another cycle is carried out [34]. This method is the most used as it is the simplest method, it can result in 0.16 to 100 microns [25].

- **Sol-gel method**

It is appropriate for making thin layers with pores; it coats the membrane with a thin layer (50 nanometer - 4 µm). In this method, the precursor sol is deposited on the support or cast into a container to get the desired shape. This method must be done in a dust-free environment to prevent defects [35].

- **Chemical vapor deposition (CVD)**

In a reaction between one or many gaseous precursors on the substrate pores, a thin film can be achieved between 400 and 1000 degrees centigrade as shown in Figure 6. This method has the benefit of eliminating the need to repeat the coating layers. Via this method, the selectivity of the membrane can be improved by optimizing the pore size and structure [35].

- **Atomic layer deposition (ALD)**

It is a new technique that could deposit thin film at a relatively low temperature compared to CVD (up to $300^{o}C$), in this method two or more precursors are reacted cyclically, the first reactant enters then purging occurs, after which the second reactant is introduced, then purging occurs to remove any by products or unreacted reactants as shown in Figure-7. This method can result in a very thin layer (in Angstroms) [35].

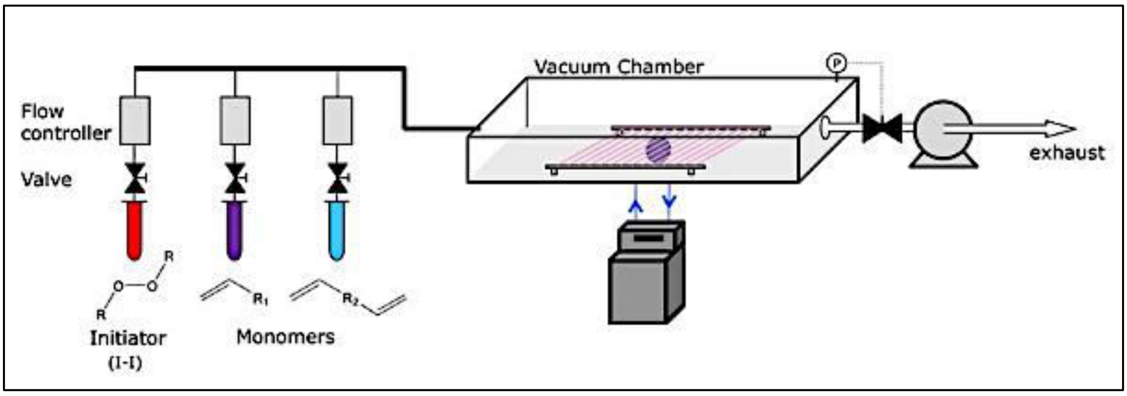

Figure-6. Schematic diagram of chemical vapor deposition [36].

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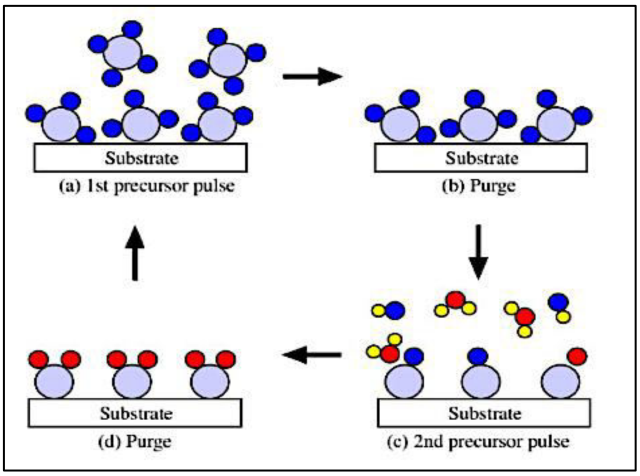

Figure-7. Steps of atomic layer deposition [37].

4. CONCLUSIONS

Membrane technology is one of the best techniques available nowadays for water treatment; ceramic membranes show promising results in addition to unique characteristics that make it one of the leading research topics. The manufacturing of ceramic membranes has different stages in which different variables are taken into consideration to produce a highly efficient and low-cost membrane. In this paper, it was concluded that:

- Ceramic membranes are promising due to their characteristics including low fouling and high lifetime, high temperature stability, and their ability to resist harsh operating conditions.
- The manufacturing of ceramic membranes includes different stages such as raw material selection, additives selection, shaping method, and choosing thermal treatment cycle and sintering temperature
- Raw materials and additives must be chosen to satisfy both the characteristics needed and low cost, it was found that the cost of one meter square of the ceramic membrane could vary from 2 up to 130$ depending on the raw materials.
- Raw materials not only affect the cost according to their price but also according to their melting point, as the thermal treatment cycle and the final sintering temperature are chosen according to it.
- Additives are important to add value to the produced ceramic membranes, such as pore forming agents, as they enhance the porosity and permeability of the membrane. On the other hand, using pore forming agents excessively could lead to several draw backs such as a decrease in the final strength.
- There are several preparation methods such as pressing, extrusion, and slip casting. Pressing is cost effective, extrusion produces membranes with high mechanical strength and high surface to volume ratio, and slip casting is simple however it is hard to control the thickness of the membrane besides needing a long time.
- The thermal treatment cycle is designed to eliminate some substances such as moisture and pore forming agents in addition to giving the membrane its final characteristics.
- It was found that by increasing the final sintering temperature the pore size, permeability, mechanical strength, and chemical resistance increase as well.
- There are several techniques for surface modification including dip coating and spraying. Chemical vapor deposition and atomic layer deposition, all these methods are effective and capable of increasing the efficiency of the membrane.
- Spray method and dip coating are used due to their ease of application and low cost.
- CVD and ALD are used to obtain thin films, ALD can achieve thin layers in the angstrom level and at lower temperatures than CVD.

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